



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPENDIX : SHOZO NAGAI, ET AL.
SERIAL NO. : 09/878,333
FILED : June 12, 2001
FOR : Ni-base brazing alloy

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O. Lawrence
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DECLARATION

Sir.

I, Shozo Nagai, a Japanese national, residing at 188 Oyake-sawacho, Yamashina-ku, Kyoto-shi, Kyoto, Japan declare:

1. I am a metallurgical engineer. I was born on December 24, 1953. I graduated from Niihama Technical College, Metallurgical Engineering Course.

2. I have been engaging in research and development on metal powder since 1974 and further in research and development on Ni-base alloy such as brazing alloys and surface hardening agents since 1987 as a research worker of Fukuda Metal Foil & Powder Co., Ltd..

3. I am a co-inventor of U.S. Patent Application Serial No.09/878,333 filed June 12, 2001 (hereinafter referred to as 'the present application'). I have studied the official action dated May 8, 2003. I have also read and studied the cited references, JP 09-225679 A, US 4,507,264 A, US 5,603,891A and US 6,203,754 B1.

4. I believe that the references do never teach any of the object, features and unexpected results of the invention defined in Claims of the present application and therefore the present invention is not obvious to those skilled in the art from the references.

5. The present invention is directed to a Ni-base brazing alloy comprising Cr in an amount of 25-35 wt.%. We, the inventors of the present application, had made investigations to overcome disadvantages of prior art such as JP 09-225679 A which is one of the cited references, and found out that an increased amount of Cr improves strength of the alloy, a specified

amount of P and Si leads formation of hypo-eutectic structure, and a certain amount of Al, Ca, Y and misch metal decreases oxygen content which in result prevents slag (Cr oxides) formation when brazed. The present invention comprising the above features, therefore, provides a brazing alloy which prevents forming slags when brazed, which attains a high strength of the brazed joint, which has a melting point as low as at about 1100 °C, and which also has good wettability, and an excellent corrosion resistance to sulfuric acid. These remarkable advantages are presented in examples summarized in Table 1 of the present specification.

6. Each of the cited references does never teach any of the object, features and unexpected results of the present invention as follows:

[JP 09-225679 A (Nagai)]

I am an inventor of the reference. The reference's invention is completed by the inventors of the present invention, Nagai, Tanaka and Hidaka, with Kwarada. Accordingly, I have well known the reference's invention.

The reference is described as a prior art having problems which must be improved at page 2 of the present specification.

The reference's invention relates to a Ni-base brazing alloy, but the alloy does never comprise any of Al, Y and the like. Resultantly the alloy can not prevent slag formation and does not attain a high strength of the brazed joint, as shown in Table 2 (a) and (b) of the present specification.

[US 6,203,754 B1 (Sugiyama)]

The reference's invention relates to a brazing filler metal whose brazing temperature is 1060 to 1120 °C and the brazing filler metal is composed of Cr, P, Si and Ni, but does not comprise any of Al, Y and the like. While the reference's invention is substantially identical to that of the above JP 09-225679 A, the priority date of the reference is January 27, 1999 which is after the publication date of the above JP 09-225679 A, namely September 2, 1997.

The alloy disclosed in the reference can not prevent slag formation and does not attain a high strength of the brazed joint, as shown in Table 2 (a) and (b) of the present specification, similarly to the case of the above JP 09-225679 A.

[US' 4,507,264 (Stern)]

The reference discloses a nickel base brazing alloy suitable for high temperature brazing and diffusion brazing of superalloys of the gamma prime type. The nickel base brazing alloy has the following composition as described at column 2, lines 7-14.

Cr	12-14 wt. %
Ta	2- 4 wt. %
B	2.5- 4 wt. %
Y or La	0.01-0.06 wt. %
Al	2.5- 5 wt. %
C	less than 0.03 wt. %
Ni	balance

The aluminum and yttrium are combined with chromium to form an improved microstructure and to improve sulfidation and oxidation of the brazed joints.

The total amount of Al and Y comprised in the alloy is 2.51-5.06 wt. % which is more than 0.1 wt. %. Further the alloy comprises Cr in an amount of 12-14 wt. % which less than 25 wt. % and does not comprise P.

Further the brazing temperature of the alloy is 2150 to 2300° F corresponding to 1176 to 1260 °C as described at column 2, line 26, and the alloy can not be used at a brazing temperature of 1100°C.

[US 5,603,891 (Brill)]

The reference relates to a heat resistant hot formable austenitic nickel alloy which mainly comprises Ni, Cr, Fe and Si but does not relates to a brazing alloy.

The austenitic nickel alloy consists of (in % by weight):

C	0.05 to 0.15
Si	2.5 to 3.0
Mg	0.2 to 0.5
P	max 0.015
S	max 0.005
Cr	25 to 30
Fe	20 to 27
Al	0.05 to 0.15
Ca	0.001 to 0.005
rare earths	0.05 to 0.15
N	0.05 to 0.20

Ni

residue

Though Al, Ca and rare earths are comprised in the alloy, they are added to improve the oxidation-resistance at temperatures in 500 to 1000°C with cyclic stressing. The total amount of Al, Ca and rare earths is 0.101 to 0.305 wt.% which is more than 0.1 wt.% and the amount of P is 0.015 wt.% or less which is very small amount in comparison with 4-8 wt. %.

Namely, Al, Y, Ca and rare earths in the reference's invention are used to improve the properties of a solid state alloy in the same manner as in US 4,507,264 (Stern), but not used to improve the properties of a melted state alloy.

As described above, each of the object of using Al, Y and the like and the used amount of US 4,507,264 (Stern) and US 5,603,891 (Brill) is very different from the present invention.

In order to clarify the inventiveness of the present invention in comparison with the reference's invention, some experimental data as shown in the following Table.

No	Composition of the alloy (wt.%)								Melting point (°C)		Brazing at 1100 °C		Transverse rupture strength (kgf/mm ²)
	Ni	Cr	P	Si	Al	Y	Al	Y	Solidus	Liquidus	Coefficient W	Slag	
Examples of the present invention													
1	Bal.	25.0	6.0	4.0	0.01	-	-	-	980	1055	50	no	84
2	Bal.	29.0	6.2	3.8	-	0.01	-	-	985	1040	50	no	94
3	Bal.	29.7	6.1	4.1	0.04	-	-	0.01	980	1025	50	no	90
4	Bal.	30.1	6.0	4.0	-	0.03	0.03	-	980	1030	50	no	91
5	Bal.	35.0	5.8	4.2	-	-	0.01	-	980	1035	50	no	86
Controls													
A	Bal.	24.6	8.3	2.8	-	-	-	-	970	1030	50	yes	55
B	Bal.	29.0	7.0	4.7	-	-	-	-	985	995	50	yes	43
C	Bal.	28.5	6.0	4.0	0.13	-	-	-	980	1020	5	no	60
D	Bal.	22.0	8.3	2.8	-	0.12	-	-	980	1040	5	no	50
E	Bal.	29.0	0.05	4.0	0.10	-	-	-	1190	1320	-	-	-

The experimental data of Nos.1-5 in above Table are same as in Nos.(1)-(5) in Table 1 of the present specification, which are according to the present invention. The experimental data of Nos.A-E are those in Controls. Nos.A-C correspond with Nos.(a)-(c) in Table 2 of the present specification. The experimental data of Nos.A and B are those obtained by the alloys described in JP 09-225679 A (Nagai) and US 6,203,754 B1 (Sugiyama). The experimental data of No.C is in the case of that Al is added in an amount of more than 0.1 wt.% to the alloy described in JP 09-225679 A (Nagai) and US 6,203,754 B1 (Sugiyama). The experimental data of No.D is about the alloy comprising more than 0.1 wt.% of Y. Further, the experimental data of No.E is about the alloy comprising P in a small amount of 0.05 wt.%.

As shown in Nos.A and B of the above Table, the alloys described in JP 09-225679 A (Nagai) and US 6,203,754 B1 (Sugiyama) which does not have such problems that a slag is formed and a strength of the brazed joint is poor. If Y, Al and the like to the alloys are added in a total amount of more than 0.1 wt.% according to the description of US 4,507,264 (Stern) and US 5,603,891 (Brill), a strength of the brazed joint can not be substantially improved and further the alloys have a low spread coefficient of melted alloy although a slag formation may be improved as shown in Nos.C and D of the above Table. Further, the brazing alloy comprising P in an amount of 0.015 wt.% or less as in US 4,507,264 (Stern) and US 5,603,891 (Brill) has a liquidus as high as more than 1200°C as shown in No.E of the above Table and accordingly can not be brazed at 1100°C. The brazing temperature of the alloys of US 4,507,264 (Stern) is 1176 to 1260°C as described above.

["Nikel, Cobalt, and their Alloys"],

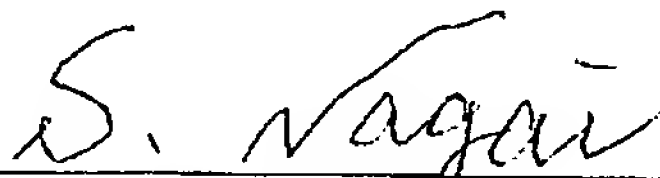
The reference describes in Table 2 at page 17 that Al is useful as an alloying element to improve oxidation and sulfidation resistance and that Y is useful as an alloying element to improve high-temperature strength, grain size control and resistance to oxidation. The effects of Al and Y are substantially identical with those described in US 4,507,264 (Stern) and US 5,603,891 (Brill) and are not necessary to attain the object of the present invention.

As described above, each of the cited references does never teach any of the object, features and advantages of the present invention.

7. I believe, therefore, that the present invention is remarkably distinguishable from the cited references and patentable.

I hereby declare that all statements made herewith of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or Imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

This 30th day of August, 2003



Shozo Nagai